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Part I. Contract Identification Information

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Part II. Completed results

In the interim report the previous investigations, devoted to the Dicke superradiance [1] and other phenomena in gamma-range, was described. It is note, that paper of J.H.Terhune and G.C.Baldwin [2] may be considered as the first theoretical investigation of the nuclear superradiance in solids. Later in paper [3] the statement "nuclear excitons" is used for the description of the gamma-superradiance. It correlates with the results of our paper [4] on exciton superradiance, in which the kinematic interaction of excitons plays the essential role. The papers [5,6], devoted to the experimental observation of gamma-echo, must be noted also. The authors of paper [7] used the statement of the collective spontaneous radiation (i.e. Dicke superradiance) for explanation of their experimental results. This statement lies in base of many papers (such as [8]), including our papers [9-11]. In paper [10] the non-Dicke mechanism of superradiance forming was grounded. In these papers [8-11], as a rule, authors considered the gamma-superradiance on the dipole nuclear transitions with the intensity, proportional to square of number of active nuclei.

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One of main results of this project is the proof of possibility of the quadrupole gamma-superradiance (QSR), the intensity of which is proportional to the forth degree of the number of active nuclei [12]. We are using the model taking into consideration the cascading character of the quadrupole transitions (E2), when the total angular momentum changes on two under the emission of one gamma-photon. This model is the modification of electrodipole Dicke model for the quadrupole transitions. We must take into consideration the three working levels: ground with the angular momentum J, intermediate - with J±1, excited - with J±2. Therefore we must introduce the effective spin S=1. Under that the operators of rise and fall of the effective spin projection correspond formally to the operators of the change of the total angular momentum of nucleus on unit. When separate nuclei quadruples are equally oriented in space as a consequence of self correlation process these operators can be substituted by relevant collective operators divided by N. So, the total hamiltonian can be written as the following

$$H = \hbar \omega_0 R_z + \hbar \omega_{\vec{k}} a_{\vec{k}}^+ a_{\vec{k}} + g_{\vec{k}} a_{\vec{k}} (R_{\vec{k}}^+)^2 + g_{\vec{k}}^* a_{\vec{k}}^+ (R_{\vec{k}}^-)^2$$
 (1)

where $\hbar\omega_0$ is an energy separation between nucleus levels, $\hbar\omega_{\vec{k}}$ - energy of gamma quantum, R_z - operator of collective population difference, $a_{\vec{k}}^+$ and $a_{\vec{k}}^-$ operators of creation and annihilation of gamma quantum with wave vector \vec{k} . This hamiltonian formally coincides with that of macroscopic quadrupole interacting with gamma quanta. The difference is that here interaction constant is normalized by N because separate quadrupoles don't form the macroscopic one by their simple coherent summation as in the case of dipoles. One example of nuclear systems with macroscopic quadrupole momentum may be clusters arising in nuclear matter with increasing density. Even-even (plus) nuclei such as 114 Ba, 150 Sm, 220 Rn, 220 Ra radiate spontaneously only on the E2-transition without electrodipole and multipole transition [13]. So, with increasing density of nuclei these systems became real candidates for observation of QSR.

Using nonequilibrium statistical operator method, we can derive from hamiltonian (1) the following kinetic equation

$$\frac{d\langle R_z \rangle}{dt} = -\frac{2}{\tau_1} \left(N + \langle R_z \rangle + N^2 - \langle R_z \rangle^2 \right)^2, \tag{2}$$

where

$$\frac{1}{\tau_1} = \frac{2\pi}{\hbar^2} \left| g_{\vec{k}} \right|^2 \delta(\omega_{\vec{k}} - 2\omega_0) \tag{3}$$

is the reversal time of isolated transition. Numerical solution of equation (2) yields the intensity of QSR

$$I = -\hbar\omega_0 \frac{d\langle R_z \rangle}{dt} \tag{4}$$

The approximate analytical solution of equation (2) writes in the vicinity of QSR peak as following

$$I = 4N^4 \frac{\hbar \omega_0}{\tau_1} \operatorname{sec} h^2 \frac{t - t_0}{2\tau_c},$$
 (5)

where $\tau_c = \tau_1 / (2N)^3$ is a correlation time and $t_0 = \tau_c N$ is a delay time.

Being measured in τ_c , the delay time is much longer than that of usual dipole superradiance. With that, correlation time is much shorter than ordinary that is $\tau_c = \tau_1/N$. But one should remember that τ_1 is independent of N only in the case of macroscopic quadrupole formation. In the case of separate quadrupole ordering, τ_1 is proportional N^2 and τ_c has the usual N dependence. So, the decay time in absolute units is always longer than in dipole case for microscopic quadrupole superradiance and always shorter for macroscopic one. Peak intensity is proportional to the fourth degree of radiating nuclei in the case of macroscopic QSR and to the second degree, as that of dipole in the case of microscopic QSR. Macroscopic QSR may be realized on the system of the paired nuclei. Mechanism of the nuclei pairing is proposed by N.N.Bogolubov [14].

Microscopic QSR has some advantages being compared with dipole superradiance. Allowed dipole transitions are very fast in gamma range. So, it is difficult to satisfy cooperation length requirements [15] and only swept-gain superradiance will be realized in practice [16]. Quadrupole transitions are much longer and these requirements can be easily satisfied.

The next main result of this project is the application of laser cooling to obtain powerful coherent pulses of gamma radiation, using the effect of quantum superradiance. The connection of Dicke superradiance and laser cooling is investigated in details in our paper

[17] (which is supported by this grant). The investigations were continued and the two main problems were analyzed: the possibility to guench relaxation processes by laser cooling and superradiance without inversion in laser cooled systems. Both stationary and dynamic regimes of laser cooling were studied.

The influence of inhomogeneous phase relaxation on superradiance was considered in many papers. Here we will employ a simple academic model developed in [18]. In accordance with this model, the superradiance intensity can be written as

$$I = -\frac{N\hbar\omega_0}{\tau_c} e^{1/T_2^*} \operatorname{sec} h^2 \left[\frac{T_2^* (1 - e^{-t/T_2^*}) - t_0}{2\tau_c} \right], \tag{6}$$

where ω_0 is the central frequency of transition, T_2^* is the time of inhomogeneous transverse relaxation, and t_0 is the integration constant. The time when the maximum of superradiance pulse is achieved is given by an approximate formula

$$t_0^* = -T_2^* \ln(1 - t_0 / T_2^*). \tag{7}$$

Consequently, the maximum intensity is written as

$$I = N \frac{\hbar \omega_0}{\tau_c} (1 - t_0 / T_2^*).$$
 (8)

The Doppler width $\delta \omega = 1/T_2^*$ that corresponds to laser cooling to the temperature T is determined in accordance with the following formula [19]:

$$\frac{1}{T_2^*} = k\Delta v = k(2k_BT/m)^{1/2}.$$
 (9)

Expressions (8) and (9) demonstrate that laser cooling provides favorable conditions for superradiance.

Laser cooling has a beneficial effect on superradiance also in the case of homogeneously broadened systems. Phenomenological consideration of homogeneous phase relaxation with relaxation time T_2 [20] yields the following formula for superradiance intensity:

$$I = N \frac{\hbar \omega_0}{4\tau_c} \left(1 - \frac{\tau_c}{T_2} \right)^2 \operatorname{sec} h^2 \left[\frac{1}{2} \left(\frac{1}{\tau_c} - \frac{1}{T_2} \right) (t - t_0) \right].$$
 (10)

Microscopic calculations performed in [21] lead to the same formula for superradiance in a crystal where dephasing due to phonon scattering is described by

$$\frac{1}{T_2} = \frac{2\pi}{\hbar^2} \sum_{\mathbf{q} \neq \mathbf{q'}} (\delta \varphi_{\mathbf{q}\mathbf{q'}}^2) \overline{\mathbf{n}}_{\mathbf{q}} (\overline{\mathbf{n}}_{\mathbf{q'}} + 1) \delta(\Omega_{\mathbf{q}} - \Omega_{\mathbf{q'}}), \tag{11}$$

where Ω_q and n_q are the frequency and the phonon number in the mode q, respectively, and $\delta \varphi$ is the constant of dipole-phonon coupling. In the case of the laser cooling of an isolated phonon mode, the number of phonons in the isolated mode is given by

$$\overline{\mathbf{n}} = \frac{\tau_1}{\tau_1} \left(\frac{\rho \Delta}{\mathbf{N}} \right) \mathbf{n}_0, \tag{12}$$

where n_0 is the initial number of phonons, ρ is the spectral density of phonons, Δ is the spectral bandwidth of the isolated mode, N is the number of impurity molecules, and τ_1 is the rate of sample heating. In the case of laser cooling we have $\bar{n} \ll n_0$. Thus, if the selected mode of resonance phonons provides the main contribution to scattering, then the formulas (10) and (11) demonstrate a beneficial effect of laser cooling on superradiance. Note that the opposite case of an adverse effect of lattice heating on superradiance was experimentally and theoretically studied in [22].

Laser cooling can be also used to ensure conditions for the detection of superradiance in dipole-dipole homogeneous broadening of the emission line because, as is well known, a microwave field, for example, can cool a dipole-dipole reservoir [23]. These concepts can be applied to gamma superradiance of nuclear excitons. In paper [24] we considered superradiance from Frenkel excitons in a molecular crystal due to nonequilibrium Bose condensation, i.e. due to spontaneous cooling. Importantly, such cooling processes not only weaken dephasing but also give rise to correlations between atoms due to the macroscopic filling of the coherent excitonic mode.

The noncoherent spontaneous emission of nucleus system can be made anisotropic by optical polarization of nuclei. Laser cooling of superfine subsystem can create conditions for gamma superradiance due to enhancement of angular and frequency selectivity of radiation process.

One can also apply laser cooling to solve the problem of population inversion by inducing effective inversion within a narrow spectral range with partial excitation of the entire inhomogeneously broadened line. Monochromatization of N_0 excited nuclei by means of laser cooling (through an optical transition in the excited state of nucleus) makes it possible to concentrate these nuclei within a narrow spectral and, possibly, spatial range, which gives rise to effective inversion. In accordance with formula (9), this effect becomes noticeable when the temperature of excited samples reaches the value of

1

$$T = \left(\frac{N_0}{N}\right)^2 \frac{1}{(kT_2^*)^2} \frac{m}{2k_B},$$

where $(T_2^*)^{-1}$ is total spectral width of all atoms. In this case, the spectral density of excited nuclei becomes equal to the equilibrium spectral density. When the recoil frequency shift exceeds the Doppler width inversionless superradiance is possible without frequency concentration since only preliminary excited atoms can interact through the radiation field because only the recoil of atom in the course of emission can keep energy and momentum conservation in subsequent absorption [25].

At the present time the preparation of the experiment for search of the coherent gamma phenomena, where the role of the collective nuclear states is large, is carried out in the gamma optics' group (laboratory of nonlinear optics; head - prof. V.V.Samartsev). The system of positrons may be probably used in our experiment. The radiative source ²⁴Na, emitting positrons, is already acquired.

Part III. Publications

- V.V.Samartsev, S.N.Andrianov. Quadrupole gamma superradiance. Proceedings of Intern. Conference "Lasers'96", Portland, USA, 1997
- 2. S.N.Andrianov, V.V.Samartsev. *Optical superradiance and laser cooling*. Laser Physics, v.7, N 1, p.314-318, 1997
- 3. S.N.Andrianov, V.V.Samartsev. *Gamma superradiance stimulated by laser cooling*. Proceedings of Intern. Workshop IGE'97, Romania, 1997
- 4. S.N.Andrianov, V.V.Samartsev. *Quadrupole gamma superradiance*. Proceedings of Intern. Workshop IGE'97, Romania, 1997

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